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13. ABSTRACT (Maximum 200 words) This Final Technical Report summarizes the most important accomplishments resulting from the work on semiconductor quantum wire and box synthesis and optical characterization carried out under the above noted grant. These accomplishments included: (1) Creation of GaAs/AlGaAs quantum wires and boxes via purely growth control on appropriately patterned mesas on GaAs(001) and GaAs(111) substrates, (2) Demonstration of their high optical quality, including the first time-resolved cathodoluminescence studies, (3) Demonstration of focused ion beam assisted Cl ₂ etching of GaAs(001) to create mesa stripes for subsequent size-reducing growth on such mesas for realization of quantum wires, (4) Demonstration of vertically self-organized growth of coherent 3D strained InAs on GaAs island quantum dots, and (5) Demonstration of the first quantum boxes laser based upon such quantum dots.				
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All Ultra-High Vacuum In-Situ Growth & Processing Approaches to Realization of
Semiconductor Nanostructure Arrays

COMBINED TECHNICAL PROGRESS
(Jan. 1, 1995 - Dec. 31, 1995)

AND

FINAL TECHNICAL REPORT
(June 1, 1993 - Dec. 31, 1996)

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April 29, 1997

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COMBINED TECHNICAL PROGRESS (Jan. 1 - Dec. 31, 1995) & FINAL TECHNICAL
REPORT (June 1, 1993 - Dec. 31, 1996)
(ARO Grant No. DAAH0493-G-0231)

I. STATEMENT OF THE PROBLEMS STUDIED:

Effort under the above referenced grant was focused upon examining some in-situ, growth controlled approaches to synthesis of semiconductor nanostructures (quantum wires and quantum boxes) and their optical behavior. To this end, efforts were made to explore two different approaches:

- (1) size-reducing growth on patterned substrates containing appropriately oriented mesas, and,
- (2) utilization of the coherent, three-dimensional(3D) islands formed in highly strained epitaxy as quantum boxes.

Studies of the first year and a half showed a greater potential of the strained 3D island quantum boxes (also dubbed self-assembled quantum dots) for optoelectronic applications. Consequently, during the second half of this contract period a greater effort was placed upon this aspect and resulted in the demonstration of the first quantum box laser made of vertically self-organized but electronically essentially uncoupled multiply stacked quantum dots as the active region. In the following we provide a brief summary of the most important results obtained in the two categories noted above. Details may be found in the publications listed in Section III.

II. SUMMARY OF THE MOST IMPORTANT RESULTS

II.1 Nanostructures via Growth on Patterned Substrates

via control of growth on non-planar patterned GaAs(111)B and GaAs(001) substrates. For GaAs(001) the system comprised of photo-lithographically patterned $\langle 100 \rangle$ oriented square mesas of linear dimensions a few microns, subsequently size-reduced to the nanoscale ($\leq 100\text{nm}$) regime via GaAs buffer layer MBE growth. On such nanoscale mesas, deposition of AlGaAs and GaAs layer(s) gave GaAs volumes confined by AlGaAs barriers. One example each of a single isolated GaAs quantum box and a multiply-confined and communicating structure is shown in the cross-sectional TEM images of figure 1, panels (a) and (b). Details are to be found in publications 1, 2, 4, 5, 6, and 9.

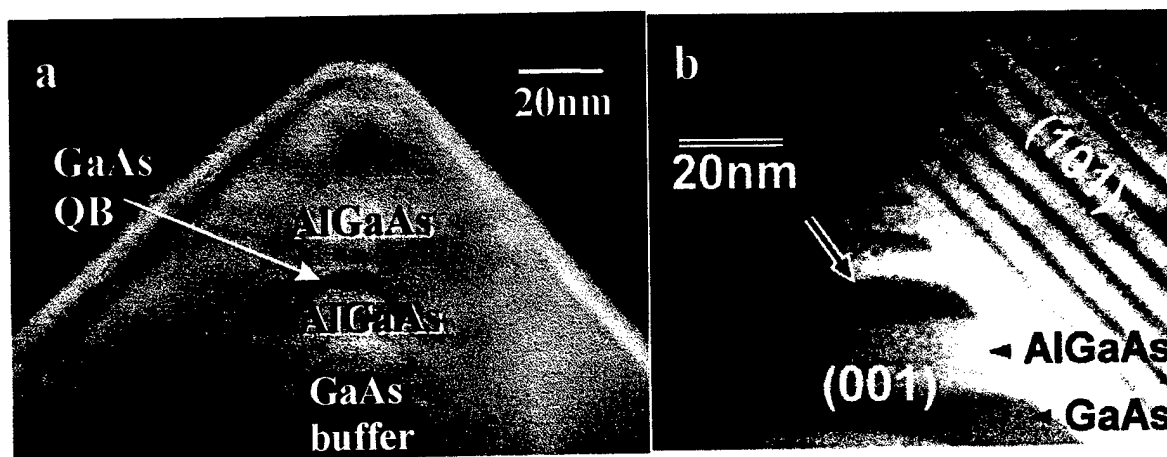


Fig. 1

2. Another important finding is the high optical quality of such structures as found from photoluminescence (PL) and cathodoluminescence (CL) studies. We note that CL studies, carried out by our colleague Prof. Daniel Rich and his students, were not called for under the original scope of this grant. However, during the course of this work, it became clear that the spatially-resolved capability of the CL would provide important optical information that would supplement and augment the large area information obtained from PL. This became even more important during the last year of our effort as Prof. Rich developed time-resolved CL capabilities, thus enabling the first space- and time-resolved CL studies of quantum boxes synthesized in-situ via growth-control. Figure 2 shows an illustrative example of the time-

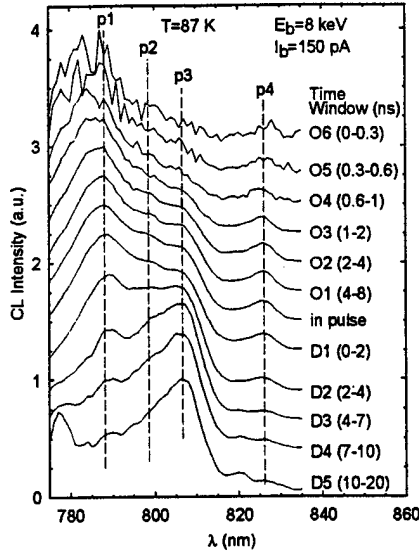


Fig. 2

oped time-resolved CL capabilities, thus enabling the first space- and time-resolved CL studies of quantum boxes synthesized in-situ via growth-control. Figure 2 shows an illustrative example of the time-resolved CL for a quantum box structure similar to that shown in fig. 1(b). Time delayed CL spectra are shown with various onset (O_i) and decay (D_i) time windows. All spectra are normalized to have about the same maximum peak height. Peak positions p1-p4 correspond to values determined from constant excitation spectra. Peaks below 800 nm are attributable to the sidewall quantum wells and peak p4 to the thick GaAs layer below the 3D confined GaAs region. Peak p3 is from the communicating GaAs confined volumes below the mesa pinch-off region. Information on carrier dynamics and state-filling effects in such growth control fabricated boxes was obtained for the first time. Details may be found in publication nos. 11, 12 and 17.

3. Additionally, some size-reducing GaAs/AlGaAs growths were carried out on in-situ prepared GaAs(001) mesa stripes. The in-situ stripe mesas oriented along $\langle 110 \rangle$ directions were prepared via focused ion beam (FIB) assisted Cl_2 etching of GaAs(001) substrates. The highly non-equilibrium nature of such FIB assisted gaseous dry etching, besides being compatible with the UHV environment of an all in-situ approach to growth/processing/re-

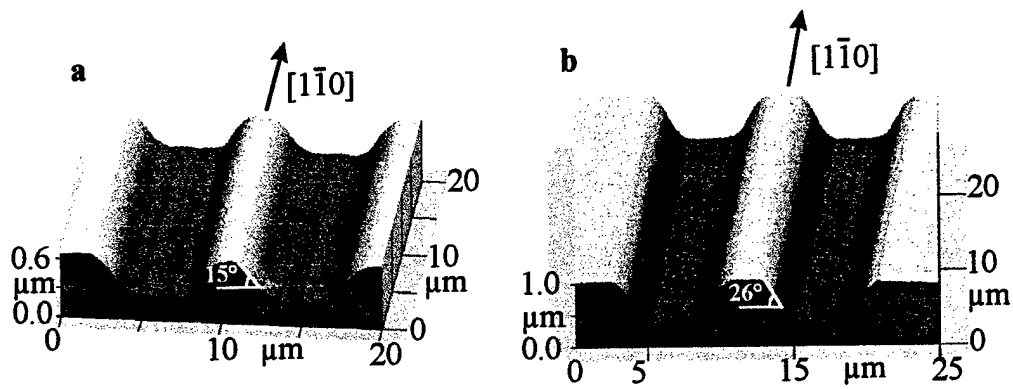


Fig. 3

shown in figs. 1 and 2. Such in-situ created mesas were characterized via atomic force microscopy (AFM). Illustrative AFM images are shown in fig. 3(a) and (b) for a very shallow ($\sim 15^\circ$) and a steeper ($\sim 26^\circ$) sidewall. Figure 4 shows the nature of the size-reducing growth on a $\sim 26^\circ$ sidewall mesa stripe as revealed by TEM images of growth of GaAs with AlGaAs (light bands) marker layers. While these results show some promising behavior, neither the scope of this grant nor the efforts demanded by the greater immediate promise of the strained 3D island quantum dots allowed us the time and resources necessary to pursue this approach much further. It is hoped that some studies can be pursued in the near future. These results are discussed in publication no. 18.

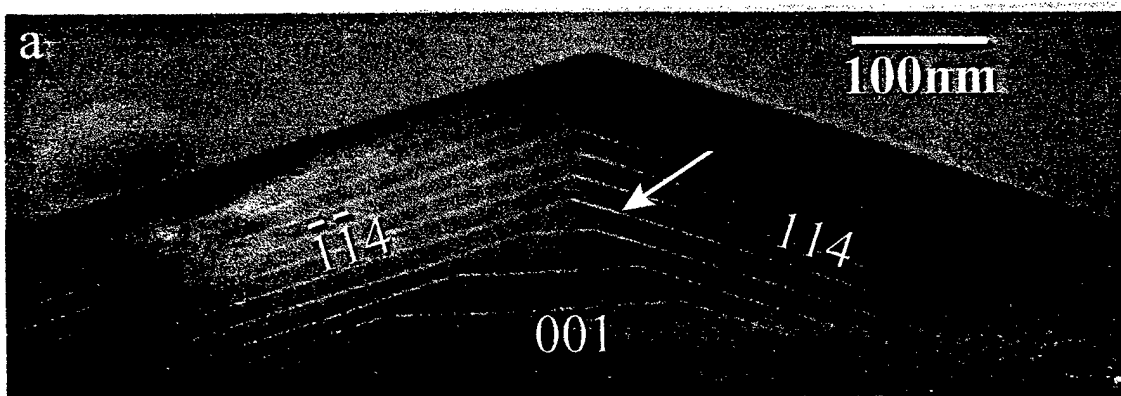


Fig. 4

II.2. Coherent, Strained 3D Island Based Quantum Dots:

The most important results obtained in this category are:

1. Demonstration of the InAs 3D island induced migration of Ga away from the islands during growth of the GaAs cap layers and the use of this phenomenon to estimate the spatial range of the island induced strain fields. This is reported in publication nos. 3 and 4.
2. The exploitation of the above noted InAs island induced stress/strain fields in GaAs cap layers to
 - (a) theoretically demonstrate the kinetically controlled occurrence of vertically self-organized 3D island stacking in multi-layer growth with varying GaAs cap(i.e. spacer) layer thicknesses, and,
 - (b) the experimental demonstration of the realization of vertically self-organized growth.

These results are to be found in publication nos. 7, 8, and 13. Figure 5 shows an illustrative cross-sectional TEM image of a stack of 5 InAs 3D island layers separated by 36 ML thick GaAs spacers and reveals the vertically self-organized growth. Figure 6 shows a comparison of the experimentally observed vertical pairing probability(symbols) and that provided by the theory based on the kinetic model (dotted line).

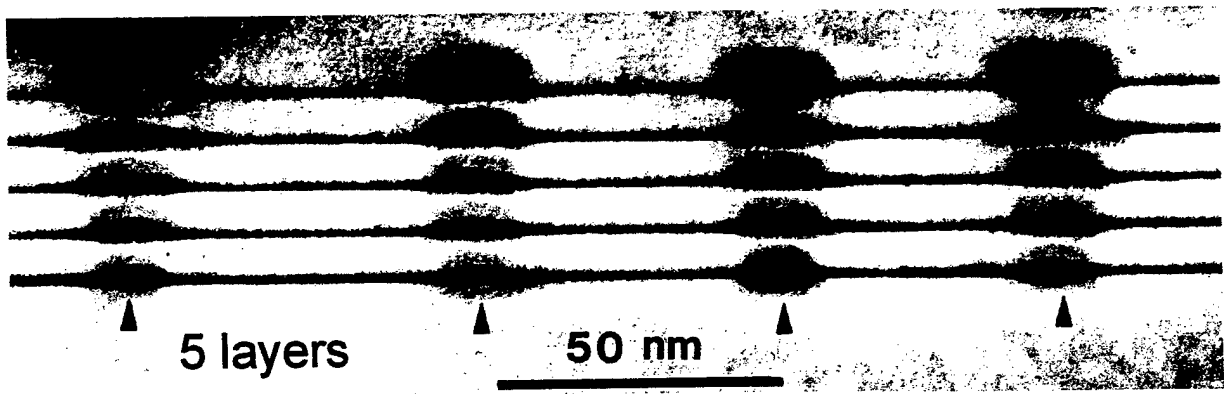


Fig. 5

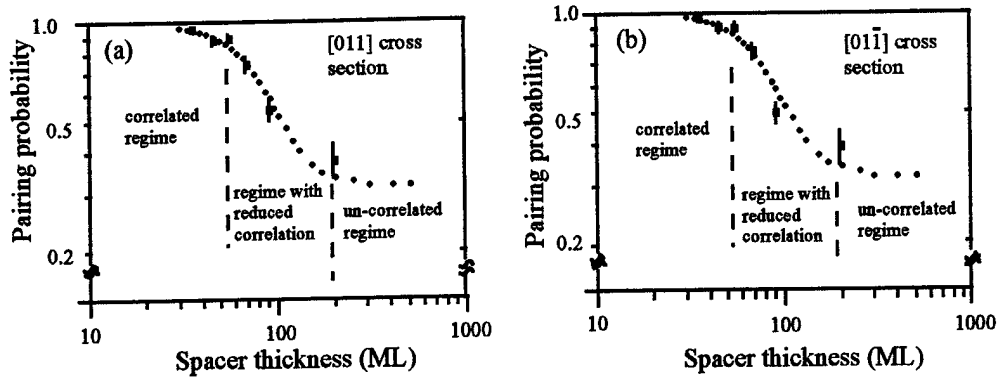


Fig. 6

3. Demonstration of Lasing from Vertically Self-Organized 3D Island Quantum Dots

Ultra low threshold lasers are a critical component of high-density, high-throughput information processing systems. Owing to the discrete density of electronic states of an ideal

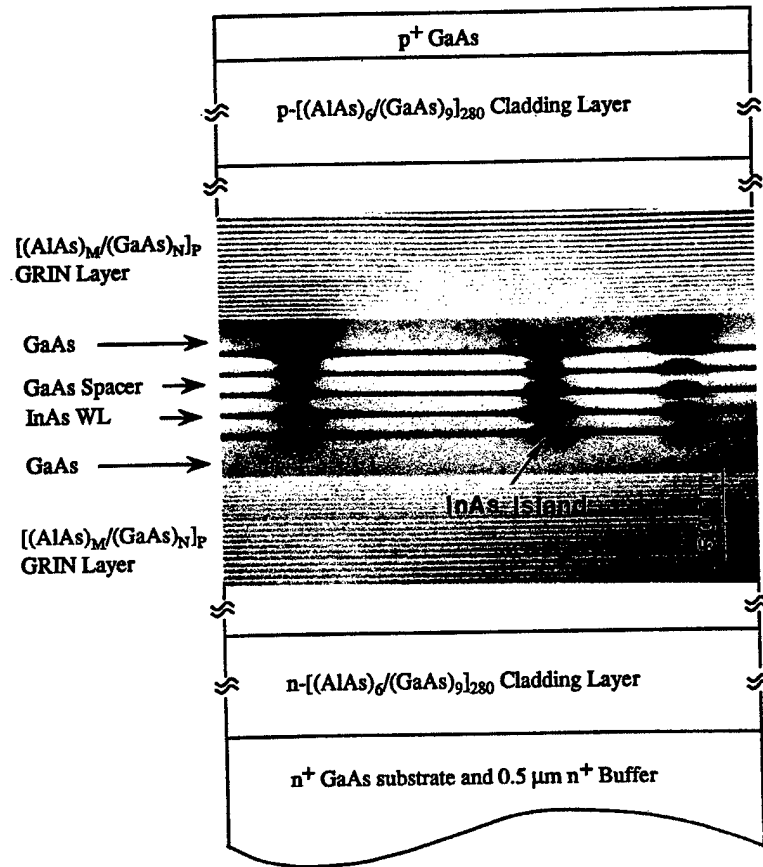


Fig. 7

quantum box, several over an order of magnitude type improvements in the figures of merit of devices based upon quantum boxes are theoretically expected. For lasers, these include threshold currents in the less than $10 \mu\text{Amp}$ regime and high characteristic temperatures leading to much improved thermal stability.

Having demonstrated the high optical quality of the singly and multiply stacked InAs 3D island quantum dots as summarized in the preceding, we embarked upon the realization and demonstration of lasing from these quantum dots. Edge emitting laser structures were designed, grown, processed, and tested. Figure 7 shows a cross-sectional TEM image of a laser structure comprising 5 sets of vertically self-organized quantum dots as the active region sandwiched between $[(\text{GaAs})_M/(\text{AlAs})_N]_P$ based graded index optical confinement layers and AlGaAs cladding layers. Figure 8 shows the light output versus injected current behavior at 77°K , indicating onset of lasing at a threshold current density (J_{th}) of $\sim 310 \text{ A/cm}^2$ (see inset), and the spectrally-resolved light output at below and above threshold. The broad lasing spectrum is a consequence of the quantum dot size fluctuations giving rise to an inhomogeneous broadening of the transitions between electron and hole states which, in turn, allow for several lasing modes to be sustained in the long cavity of an edge-emitting laser. Further details are provided in publication no. 16.

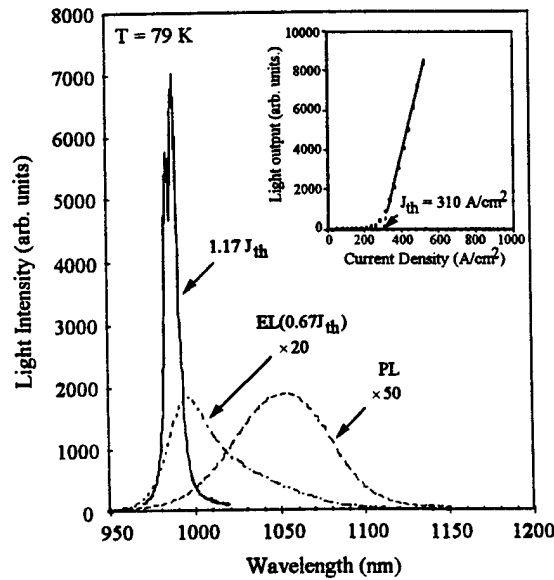


Fig. 8

Finding growth control approaches to reduce the 3D island quantum dot size fluctuation from its present best of ~10%, and finding means of achieving spatially regular 2D and 3D arrays, are the most important quantum dot synthesis control issues that deserve to be pursued in light of the potential of quantum dots demonstrated by our and recent other studies. We finally observe that the real potential of quantum dot based ultra low threshold lasers lies in vertical cavity surface-emitting lasers (VCSELs) which have just begun to be demonstrated.

III. LIST OF PUBLICATIONS

1. K.C. Rajkumar, A. Madhukar, P. Chen, A. Konkar, L. Chen, K. Rammohan, and D. Rich, "Realization of Three-Dimensionally Confined Structures via One-Step In-Situ MBE on Appropriately Patterned GaAs(111)", J. Vac. Sci. Technol. **B12**, 1071(1994)
2. A. Konkar, K.C. Rajkumar, Q. Xie, P. Chen, A. Madhukar, H.T. Lin, and D.H. Rich, "*In-Situ* Fabrication of Three-Dimensionally Confined GaAs and InAs Volumes via Growth on Nonplanar Patterned GaAs(001) Substrates", J. Cryst. Growth **150**, 311(1995)
3. Q. Xie, P. Chen, A. Kalburge, A. Nayfanov, T.R. Ramachandran, A. Konkar and A. Madhukar, "Realization of Optically Active Strained InAs Island Quantum Boxes on GaAs (100) via MBE and the Role of Island Induced Strain Fields", J. Cryst. Growth **150**, 357(1995)
4. Q. Xie, A. Konkar, A. Kalburge, T.R. Ramachandran, P. Chen, R. Cartland, A. Madhukar, H.T. Lin and D.H. Rich, "Structural and Optical behavior of Strained InAs Quantum Boxes Grown on Planar and Patterned GaAs(100) Substrates by Molecular Beam Epitaxy", J. Vac. Sci. Technol. **B13**, 642(1995)
5. A. Madhukar, P. Chen, Q. Xie, A. Konkar, T.R. Ramachandran, N.P. Kobayashi, and R. Viswanathan, "Semiconductor Nanostructures: Nature's Way", in "Low Dimensional Structures prepared by Epitaxial Growth or Regrowth on Patterned Substrates", eds. K. Eberl et al, (Kluwer Academic Publishers, Netherlands, 1995) p. 19-23
6. A. Konkar, A. Madhukar, and P. Chen, "Creating Three-Dimensionally Confined Nanoscale Strained Structures via Substrate Encoded Size-Reducing Epitaxy and the Enhancement of

Critical Thickness for Island Formation", Paper presented at MRS Spring '95 Meeting (Apr. 17-21, 1995, San Francisco, CA), MRS Symposium Proc. Vol. **380**, 17-22 (1995)

7. Q. Xie, N.P. Kobayashi, T.R. Ramachandran, A. Kalburge, P. Chen, and A. Madhukar, "InAs Island Quantum Box Formation and Vertical Self-Organization on GaAs(100) via Molecular Beam Epitaxy", Paper presented at MRS Spring '95 Meeting (Apr. 17-21, 1995, San Francisco, CA), MRS Symposium Proc. Vol. **379**, 177-183 (1995)
8. Q. Xie, A. Madhukar, P. Chen and N.P. Kobayashi, "Vertically Self-Organized InAs Quantum Box Islands on GaAs(100)", Phys. Rev. Lett. **75**, 2542-2545 (1995)
9. A. Madhukar, A. Konkar, and P. Chen, "In-Situ Formation of Laterally Confined Semiconductor Structures via Growth on Nonplanar Patterned Substrates", in "Fabrication and Characterization of Advanced Materials", eds. S. W. Kim and S. J. Park, (The Materials Research Society of Korea, 1995), 1173-1181
10. A. Madhukar, W. Yu, R. Viswanathan and P. Chen, "Some Computer Simulations of Semiconductor Thin Film Growth and Strain Relaxation in a Unified Atomistic and Kinetic Model", Paper presented at MRS Fall '95 Meeting (Nov. 27 - Dec. 1, 1995, Boston, MA), MRS Symposium Proc. Vol. **408**, 413-425 (1995)
11. D.H. Rich, H.T. Lin, A. Konkar, P. Chen, and A. Madhukar, "Time-Resolved Cathodoluminescence Study of Carrier Relaxation in GaAs/AlGaAs Layers Grown on a Patterned GaAs(001) Substrate", Appl. Phys. Lett. **69**, 665-667 (1996)
12. H.T. Lin, D.H. Rich, A. Konkar, P. Chen and A. Madhukar, "Carrier Relaxation and Recombination in GaAs/AlGaAs Quantum Heterostructures and Nanostructures Probed with Time-Resolved Cathodoluminescence", J. Appl. Phys. **81**, 3186-3195 (1997)
13. Q. Xie, N.P. Kobayashi, R.R. Ramachandran, A. Kalburge, P. Chen and A. Madhukar, "Strained Coherent InAs Quantum Box Islands on GaAs(100): Size Equalization, Vertical Self-Organization, and Optical Properties", Journal Vacuum Science Technology **B14**, 2203-2207 (1996)
14. A. Madhukar, "A Unified Atomistic and Kinetic Framework for Growth Front Morphology Evolution and Defect Initiation in Strained Epitaxy", J. Cryst. Growth **163**, 149-164 (1996)

15. N.P. Kobayashi, T.R. Ramachandran, P. Chen, and A. Madhukar, "In-Situ, Atomic Force Microscope Studies of the Evolution of InAs Three-Dimensional Islands on GaAs(001)", Appl. Phys. Lett. **68**, 3299-3301(1996)
16. Q. Xie, A. Kalburge, P. Chen, and A. Madhukar, "Observation of Lasing from Vertically Self-Organized InAs Three-Dimensional Island Quantum Boxes on GaAs(001)", IEEE Photonics Technology Letters, Vol. **8**, 965-967(1996)
17. D.H. Rich, H.T. Lin, A. Konkar, P. Chen, and A. Madhukar, "Cathodoluminescence Study of Band Filling and Carrier Thermalization in GaAs/AlGaAs Quantum Boxes", J. Appl. Phys. **81**, 1781 (1997)
18. A. Kalburge, A. Konkar, T.R. Ramachandran, P. Chen, and A. Madhukar, "Focused Ion Beam Assisted Chemically Etched Mesas on GaAs(001) and the Nature of Subsequent Molecular Beam Epitaxial Growth", J. Appl. Phys. (In Press)
19. T.R. Ramachandran, R. Heitz, N.P. Kobayashi, A. Kalburge, W. Yu, P. Chen, and A. Madhukar, "Re-Entrant Behavior of 2D to 3D Morphology Change and 3D Island Lateral Size Equalization via Mass Exchange in Stranski-Krastanow", J. Cryst. Growth (In Press)

IV. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL

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	A. Konkar	Ph. D. (1997 Dec. Expected)
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	Prof. Ping Chen	

V. REPORT OF INVENTIONS: None